NUCLEAR DATA AND MEASUREMENTS SERIES

# ANL/NDM-18

# The Delayed Neutron Yield of $^{238}$U and $^{241}$Pu

by

J.W. Meadows

January 1976

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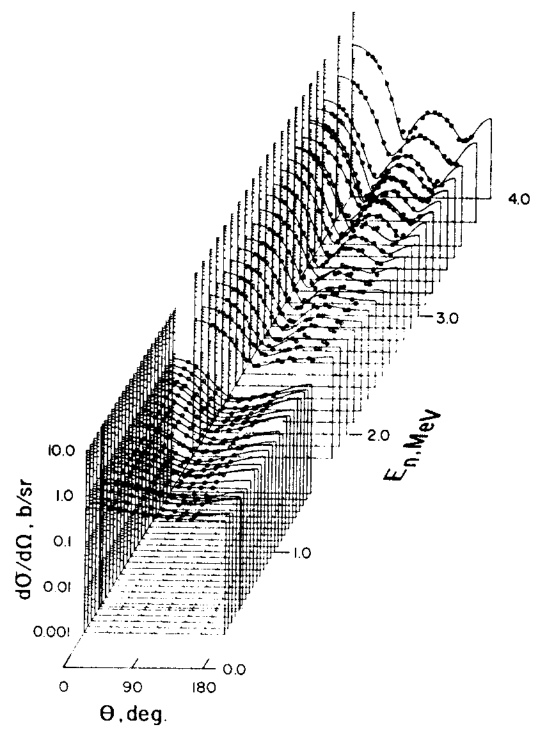


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In January 1975, the research and development functions of the former U.S. Atomic Energy Commission were incorporated into those of the U.S. Energy Research and Development Administration.

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## NUCLEAR DATA AND MEASUREMENTS SERIES

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# The Delayed Neutron Yield of $^{238}U$ and $^{241}Pu$*

by

J. W. Meadows

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## ABSTRACT

The total delayed neutron yield for $^{238}U$ and $^{241}Pu$ were observed as a function of the incident neutron energy. The measurements extend from 2.5 to 5 MeV for $^{238}U$ and from 0.15 to 5 MeV for $^{241}Pu$. The average ratio of the $^{241}Pu$ delayed neutron yield to that of $^{238}U$ is 0.292 ± .022.

*This work was performed under the auspices of the U.S. Energy Research and Development Administration.

# I. INTRODUCTION

A knowledge of delayed neutron yields from neutron induced fission is essential to the understanding of reactor kinetics and, with the advent of the fast breeder, delayed neutron data for the plutonium isotopes are of increasing importance. Recent surveys of the available data by Cox[1] and Tuttle[2] show that the delayed neutron yield of $^{239}$Pu is well determined below 4 MeV incident neutron energy but there are few measurements for the heavier isotopes. The only $^{241}$Pu measurement below 4 MeV was by Cox[3] who reported 0.0154 ± .0015 for thermal neutron fission.

Another important isotope is $^{238}$U. Krick and Evans[4] have shown that the delayed yield is independent of energy below 5 MeV, and the surveys show a number of measurements in this energy range. The results are widely scattered. Absolute measurements[1,5-8] range from 0.037 to 0.0484 and relative measurements[9,10] are also in poor agreement. The data falls into two groups as illustrated in Fig. 2. If the adjusted values given in Reference 2 are used the higher group[7-9] averages 0.0477 while the lower group[1,6,10] averages 0.0436.

This experiment was undertaken when three $^{241}$Pu samples, originally intended for transmission measurements, became available. The two heavier samples were enclosed in an aluminium capsule and used to measure the delayed neutron yield and its energy dependence. The initial measurements on $^{238}$U were made in order to become familiar with the experimental method. The large delayed neutron yield, availability, and ease of handling made it ideal for this purpose. The $^{238}$U measurements were later extended to several samples of different dimensions in order to check the correction procedures.

This paper reports the current status of these measurements.

## II. EXPERIMENTAL METHOD AND PROCEDURES

The basic technique was similar to that developed by Masters, Thorpe and Smith[7]. A fissionable sample was bombarded by neutrons from a source modulated on and off with a period much less that the half-life of any known delayed neutron precursor. After irradiation had continued for a time long compared to the longest precursor half-life the delayed neutron emission rate reached an equilibrium value which was essentially independent of time. The delayed neutrons were then counted by a detector operating in the intervals when the source was off.

The equipment used was originally developed by Cox[11] and is shown schematically in Fig. 1. Neutrons generated by the $^{7}Li(p,n)^{7}Be$ reaction caused fissions in the sample and the delayed neutrons were detected in the $^{10}BF_3$ counter assembly. A fission chamber directly behind the sample monitored the neutron flux. The proton beam was accelerated by the Argonne Tandem Dynamitron facility. The beam was modulated by deflection plates in the injector or at the exit of the accelerator. Both methods worked equally well. Measurements were taken at source to monitor distances of 2.59, 3.86, and 5.13 cm using two timing sequences. In one the sample was bombarded for 4.428 msec. A delay of 2.644 msec permitted the $^{10}BF_3$ counters to recover. The delayed neutrons were then counted for 4.428 msec. After an additional 0.5 msec the cycle was repeated. In the second sequence the corresponding intervals were 35.55, 7.00, 35.55 and 3.50 msec. Background counts were made in a similar manner but with the sample removed. Spontaneous fission neutrons were counted with the sample in place and the neutron source turned off.

The neutron detector was an array of $BF_3$ counters in a polyethylene moderator, as shown in Fig. 1. The detector stability was checked several times a day by counting a Pu-Be($\alpha$,n) source in a standard geometry.

The diameters and weights of the samples are listed in Table III and the isotopic analyses are given in Table I. The $^{241}$Pu sample consisted of two disks enclosed in an aluminum capsule with 0.025 cm thick ends and 0.13 cm thick sides. The samples were originally designed for transmission measurements and did not have the optimum dimensions for delayed neutron studies. As a result the corrections for fissions by scattered neutrons and for neutron multiplication were large. The sample originally contained 93.7% $^{241}$Pu but at the time of the measurement the $^{241}$Am content had grown to 7.2%. The $^{238}$U samples were made from depleted uranium. One of them (U-238-4) was about the size of the $^{241}$Pu sample and was enclosed in a similar capsule.

The neutron flux monitor was an ion chamber containing 2.54 cm dia. deposits of $^{235}$U or $^{238}$U. The chamber was lightly constructed to reduce neutron scattering. The two types of deposits were used because some of the correction factors discussed in the next section were significantly different, but the masses were not independently determined. The mass of the $^{235}$U deposit was measured in a previous experiment[12] and was confirmed to within 1% by comparing the alpha decay rate with that calculated from the half-lives given in Table II. The mass of the $^{238}$U deposits were obtained by comparing their fission rates at 2.5 MeV to several $^{235}U$ deposits. The method is described elsewhere[12]. Three monitor deposits were used; a $^{235}U$ with 81.3 μg uranium/cm$^2$ and two $^{238}U$ deposits with 501 and 364 μg/cm$^2$.

## III. DATA ANALYSIS

The delayed neutron yield in terms of delayed neutrons per fission in the sample is

$$N_d = \left[\frac{m\ C_d}{\varepsilon\ g\ T_c}\right] \left[C_m\ f_m\right]\ R^{-1}$$

$C_d$ = number of delayed neutron detector counts.

$m$ = multiplication and capture factor for delayed neutrons in the sample.

$\varepsilon$ = neutron detector efficiency.

$g$ = duty cycle of the delayed neutron detector.

$T_c$ = transmission and scattering factor for delayed neutrons in the fission chamber.

$C_m$ = number of monitor counts.

$f_m$ = correction factor for loss of monitor counts in uranium deposits and for pulses below the discriminator level.

$R$ = the number of fissions in the sample per monitor fission.

The principal problem is the determination of R. It may be readily calculated for the samples where scattering and multiplication is negligible but this is rarely the case in practice where a significant fraction of the fissions in both sample and monitor are due to scattered and fission neutrons. R may be expressed as

$$R = \frac{f^x_{th}\ \sum_i G_i\ N^x_i}{f^m_{th}\ \sum_i G_i\ N^m_i}$$

The superscripts x and m refer to sample and monitor respectively and

$$N_i = (D_i + S_i + M_i)T_{si}$$

$f_{th}$ = factor to account for fissions due to thermal and other room return neutrons,

$G_i$ = fraction of incident neutron associated with the $i^{th}$ neutron group,

$D_i$ = first collision fissions in the isolated sample or monitor per source neutron,

$S_i$ = fissions produced by scattered neutrons per source neutron,

$M_i$ = fissions caused by prompt fission neutrons from the sample per source neutron,

$T_{si}$ = transmission coefficient for sample and fission chamber wall

If the sample is a single isotope then Eqs. (1) and (2) give the true delayed neutron yield. However, the $^{241}Pu$ sample is a mixture of isotopes so additional corrections are required. The observed delayed neutron yield for relatively thin samples can be expressed as

$$N_d = \sum_i \frac{G_i}{\langle\sigma^x\rangle_i} \sum_j \langle\sigma^j\rangle_i \, Y_{ij} F_j \qquad (3)$$

$\langle\sigma^x\rangle_i$ = the fission cross section of the sample averaged over neutron group i.

$\langle\sigma^j\rangle_i$ = the fission cross section of isotope j averaged over neutron group i.

$Y_{ij}$ = the delayed neutron yield of isotope j averaged over neutron group i.

$F_i$ = the isotopic fraction of i.

Eq. (3) can be readily solved for a particular $Y_{ij}$.

The scattering and multiplication corrections were calculated by a Monte Carlo procedure and include elastic and inelastic scattering by the sample, sample capsule, the lithium target support plate, and the fission chamber. Total, elastic, inelastic and fission cross sections were taken from ENDF/B-IV[16]. Experimental angular distributions[17] were used for elastic scattering but all inelastic scattering was assumed to be isotropic. The delayed neutron yields of $^{239}$Pu, $^{240}$Pu, $^{242}$Pu, and $^{241}$Am were assumed to be independent of energy. Experimental values were used for $^{239}$Pu and $^{240}$Pu but the $^{241}$Am and $^{242}$Pu values were based on systematics [1,2]. The values used are listed in Table I. Three neutron groups were considered in Eq. (2). These were $^{7}$Li(p,n)$^{7}$Be (g.s.), $^{7}$Li(p,n)$^{7}$Be$^{*}$, $^{7}$Li(p,n$^{3}$He)$^{4}$He[18]. A scattered neutron group and a fission neutron group were added for Eq. (3).

Most of the remaining correction factors were determined experimentally. The room return factor, $f_{th}$, was due to thermal or near thermal neutrons and was negligible for $^{238}$U. It was measured by pulsed beam and fast timing techniques and ranged from 1.01 to 1.03 for $^{235}$U, depending on the neutron source to sample distance. The monitor counts correction factor, $f_m$, accounted for counts lost below the discriminator level and for fission fragments that were emitted at such large angles they did not emerge from the deposit. The correction was obtained by extrapolating the fission spectrum to zero and by measuring the specific fission rates for a series of uranium deposits ranging from 40 to 500 $\mu g/cm^2$. The factors ranged from 1.029 for the $^{235}$U deposit to 1.157 for the thicker $^{238}$U deposit. However only the $^{235}$U factor was significant. Since the $^{238}$U masses were based on their fission rates relative to $^{235}$U their $f_m$ factors canceled. The delayed neutron transmission factor was calculated. It was very nearly 1.0 since the in- and out-scattering almost canceled.

Some typical values of the correction factors are listed in Table III. Their dependence on sample dimensions emphasize the desirability of choosing these dimensions to minimize the effects of scattering and multiplication. Table IV shows the corresponding delayed neutron yields for $^{238}U$. There may be some dependence on sample size, but the effect, if any, is comparable to the uncertainty in the measurements. The generally good agreement is evidence of the reliability of the scattering corrections.

## IV. SOURCES OF ERROR

A summary of the estimated errors is given in Table IV. The $^{241}Pu$ results are based on $^{235}U$ and $^{238}U$ monitors so there is a 5% error which is essentially the error in the ratio of the $^{241}Pu$ to $^{235}U$ fission cross sections. The $^{238}U$ delayed neutron yields was measured with a $^{238}U$ monitor so there is no problem with relative fission cross sections. The $^{238}U$ to $^{235}U$ fission cross section was used in measuring the mass of the $^{238}U$ deposits, but that error is small[12] and is included in the deposit weight error. The error due to the uncertainty in the relative intensities of the neutron groups are small because the fission cross sections and delayed neutron yields have little energy dependence. All the errors were added quadratically. Errors due to uncertainties in the delayed neutron spectra and in the energy dependence of the detector efficiency have not been established so they are not included.

## V. RESULTS

Only relative values are reported due to uncertainties in the energy dependence of the delayed neutron detector. The average of all measurements given in Table IV was normalized to the $^{238}U$ value of Masters et al.[7] as modified in Reference 2.

The energy dependence of the delayed neutron yields are shown in Fig. 2. Krick and Evans[4] have shown that the delayed neutron yield for a number of isotopes (including $^{238}U$ but not $^{241}Pu$) is independent of energy below 4 or 5 MeV. The $^{238}U$ results are in good agreement with their observations. The behavior of $^{241}Pu$ is similar above 1 MeV but the delayed neutron yield shows a decided peak near 0.3 MeV. It cannot be explained as a statistical anomaly. The relative errors are too small (3-4%) and the behavior is too systematic. Still there is no readily apparent reason to expect $^{241}Pu$ to exhibit an energy dependence that differs from $^{235}U$ or $^{239}Pu$.

This apparent energy dependence may actually be a reflection of errors in the $^{241}Pu$ to $^{235}U$ fission cross section ratios. A number of measurements have been reported.[19-12] Käppeler and Pfletschinger[21] have extensive measurements below 1 MeV. Smith,[20] Smith and Nobles[23] have data from 0.12 to 21 MeV but their values below 1 MeV are larger than those of Käppeler and Pfletschinger by as much as 10%. Fission cross section ratios calculated from the ENDF/B-IV data set tend to follow the results of Smith et al. above 1.5 MeV and those of Käppeler and Pfletschinger below 1 MeV. Thus it is very probable that the ENDF/B-IV ratios are in error as far as shape is concerned. When the $^{241}Pu$ delayed neutron yields are calculated using the $^{241}Pu$ to $^{235}U$ fission cross section ratios of Smith et al. the peak near 0.3 MeV disappears.

Recently Behrens and Carlson[24] reported preliminary results for the $^{241}Pu$ to $^{235}U$ fission cross section ratios from 0.1 to 30 MeV. These are larger than the ENDF/B-IV values near 0.3 MeV and smaller above 1.5 MeV. When the $^{241}Pu$ delayed neutron yields are calculated using these results the peak near 0.3 MeV is no longer significant.

If the delayed neutron yields have no energy dependence below 4-5 MeV all data in that energy range may be averaged. When this is done for $^{241}Pu$ those values are obtained depending on the set of $^{241}Pu$ to $^{235}U$ fission cross section ratios used to calculate the yields. They are

| | |
|---|---|
| ENDF/B-IV | 0.0142 ± .0011 |
| Smith et al. | 0.0137 ± .0011 |
| Behrens et al. | 0.0145 ± .0009 |

The uncertainties do not include the normalization error. If the ENDF/B-IV result is used the ratio of the $^{241}Pu$ delayed neutron yield to that of $^{238}U$ is

$$Y_d(^{241}Pu)/Y_d(^{238}U) = 0.292 \pm .022$$

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TABLE I

Isotopic Composition of the Samples and the Delayed Neutron Yields of the Contaminants

| Isotope | Atom % | | Delayed Neutron Yield |
|---|---|---|---|
| | U-238 Sample | Pu-241 Sample | |
| U-235 | 0.22 | | 0.0170[a] |
| U-238 | 99.78 | | |
| Pu-239 | | 0.6 | 0.0066[a] |
| Pu-240 | | 3.0 | 0.0096[a] |
| Pu-241 | | 86.5 | |
| Pu-242 | | 2.7 | 0.024[b] |
| Am-241 | | 7.2 | 0.0045[b] |

a. Reference 2.

b. Estimated from systematics. References 1 and 2.

TABLE II

Composition and Half Lives for the Uranium Monitor Deposits

| Isotope | Atom % | | Half Life in Years |
|---|---|---|---|
| | U-235 Monitor | U-238 Monitor | |
| U-234 | 0.856 | | $2.455 \times 10^5$ [a] |
| U-235 | 93.249 | 6 ppm | $7.038 \times 10^8$ [b] |
| U-236 | 0.332 | | $2.342 \times 10^7$ [c] |
| U-238 | 5.526 | 100.00 | $4.468 \times 10^9$ [b] |

a. Ref. 13

b. Ref. 14

c. Ref. 15

TABLE III

Neutron Scattering and Multiplication Corrections at 2.5 MeV

| Sample | U-238-1 | U-238-2 | U-238-3 | U-238-4 | Pu-241 |
|---|---|---|---|---|---|
| Diameter, cm | 2.540 | 2.540 | 2.540 | 1.688 | 1.688 |
| Weight, g. | 9.897 | 19.58 | 48.26 | 15.31 | 11.46 |
| $S_1^x/D_1^x$ | 0.066 | 0.092 | 0.155 | 0.128 | 0.157 |
| $M_1^x/D_1^x$ | 0.009 | 0.014 | 0.028 | 0.022 | 0.077 |
| m | 1.0 | 0.998 | 0.997 | 0.997 | 1.052 |
| $f_{th}^x$ | 1.0 | 1.0 | 1.0 | 1.0 | 1.01 |
| Monitors | | | | | |
| $T_{s1}$ | 0.947 | 0.912 | 0.816 | .93 | .95 |
| U-238 | | | | | |
| $S_1^m/D_1^m$ | 0.055 | 0.075 | 0.123 | 0.059 | 0.082 |
| $M_1^m/D_1^m$ | 0.004 | 0.008 | 0.015 | 0.007 | 0.009 |
| $f_{th}^m$ | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| U-235 | | | | | |
| $S_1^m/D_1^m$ | 0.075 | 0.102 | 0.169 | | 0.086 |
| $M_1^m/D_1^m$ | 0.008 | 0.014 | 0.030 | | 0.022 |
| $f_{th}^m$ | 1.01 | 1.02 | 1.03 | | 1.01 |

TABLE IV

Delayed Neutrons Per Fission for 2.5 MeV Neutrons Incident on U-238

| Sample No | | 1 | 2 | 3 | 4 | Average |
|---|---|---|---|---|---|---|
| Monitor | Distance cm | | | | | |
| U-238-60 | 2.540 | .0497 | .0504 | | .0466 | .0489 |
| | 3.81 | .0477 | .0477 | .0471 | | .0475 |
| | 5.08 | | .0502 | .0469 | | .0486 |
| U-238-59 | 3.81 | .0512 | .0521 | .0502 | | .0512 |
| U-235-5 | 3.81 | .0492 | .0475 | .0468 | | .0478 |
| Average | | .0494 | .0496 | .0478 | .0466 | |

TABLE V

Summary of Errors

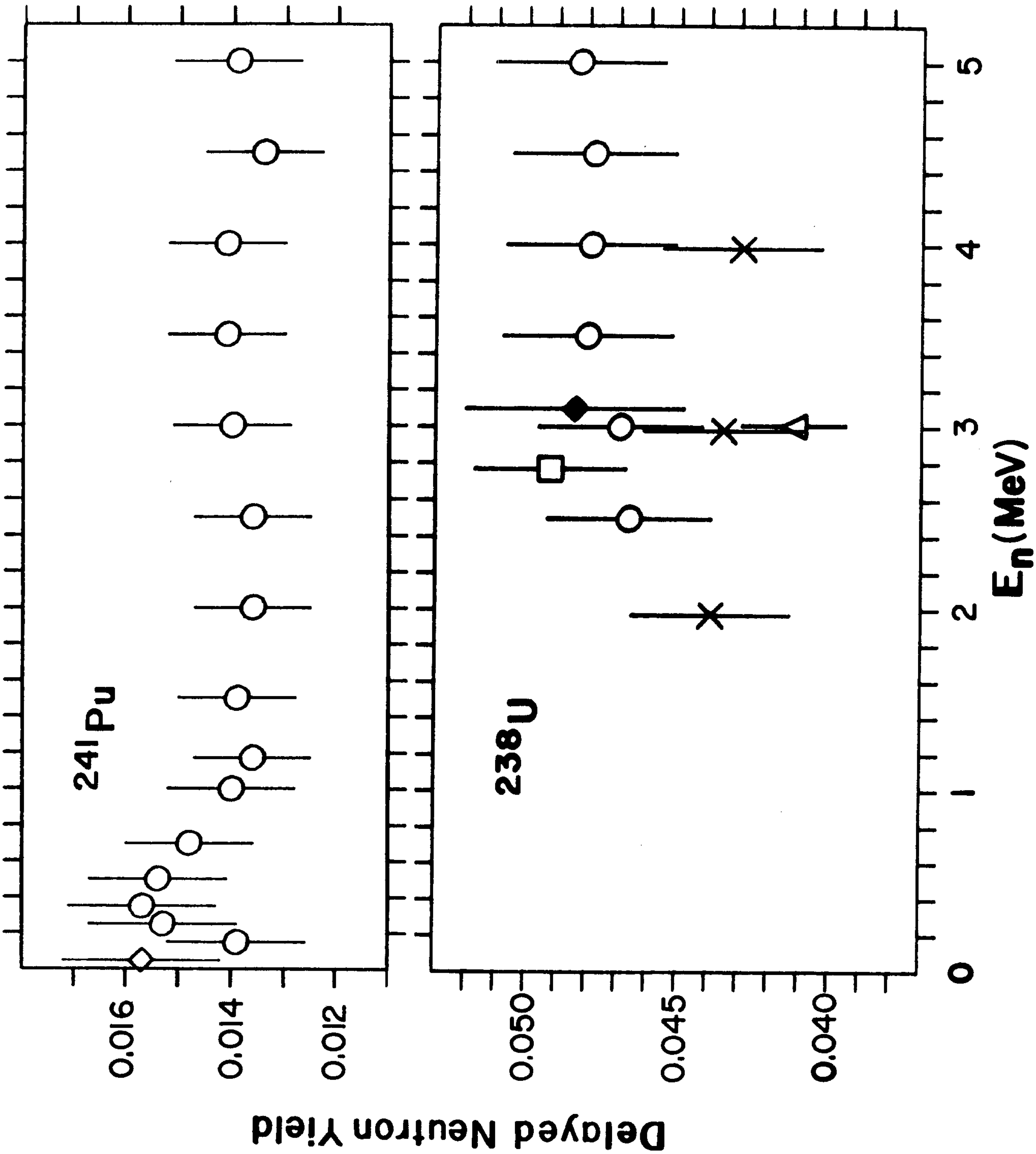
| | $^{238}U$ | $^{241}Pu$ |
|---|---|---|
| Statistical | 1-2 % | 1-2 % |
| Scattering Correction | 1-5 | 2-3 |
| Monitor Deposit Weight | 2 | 2 |
| Relative Fission $\sigma$ | 0 | 5 |
| Dimensional Errors | 2 | 5 |
| Total | 3-6 % | 7-8 % |

## FIGURE CAPTIONS

Fig. 1. A schematic diagram of the experimental setup.

Fig. 2. The $^{238}U$ and $^{241}Pu$ delayed neutron yields. The $^{238}U$ results are for sample No. 4. The data is normalized to the $^{238}U$ delayed neutron yield of Masters et al.[7] as described in the text. The open circles (O) are the results of this experiment, (◆) reference 7, (□) reference 8, (△) reference 6 (X) reference 1, and (◇) reference 3.

Fig. 1.



$^{241}Pu$
$^{238}U$
Delayed Neutron Yield
0.016
0.014
0.012
0.050
0.045
0.040
$E_n$ (MeV)
0
1
2
3
4
5

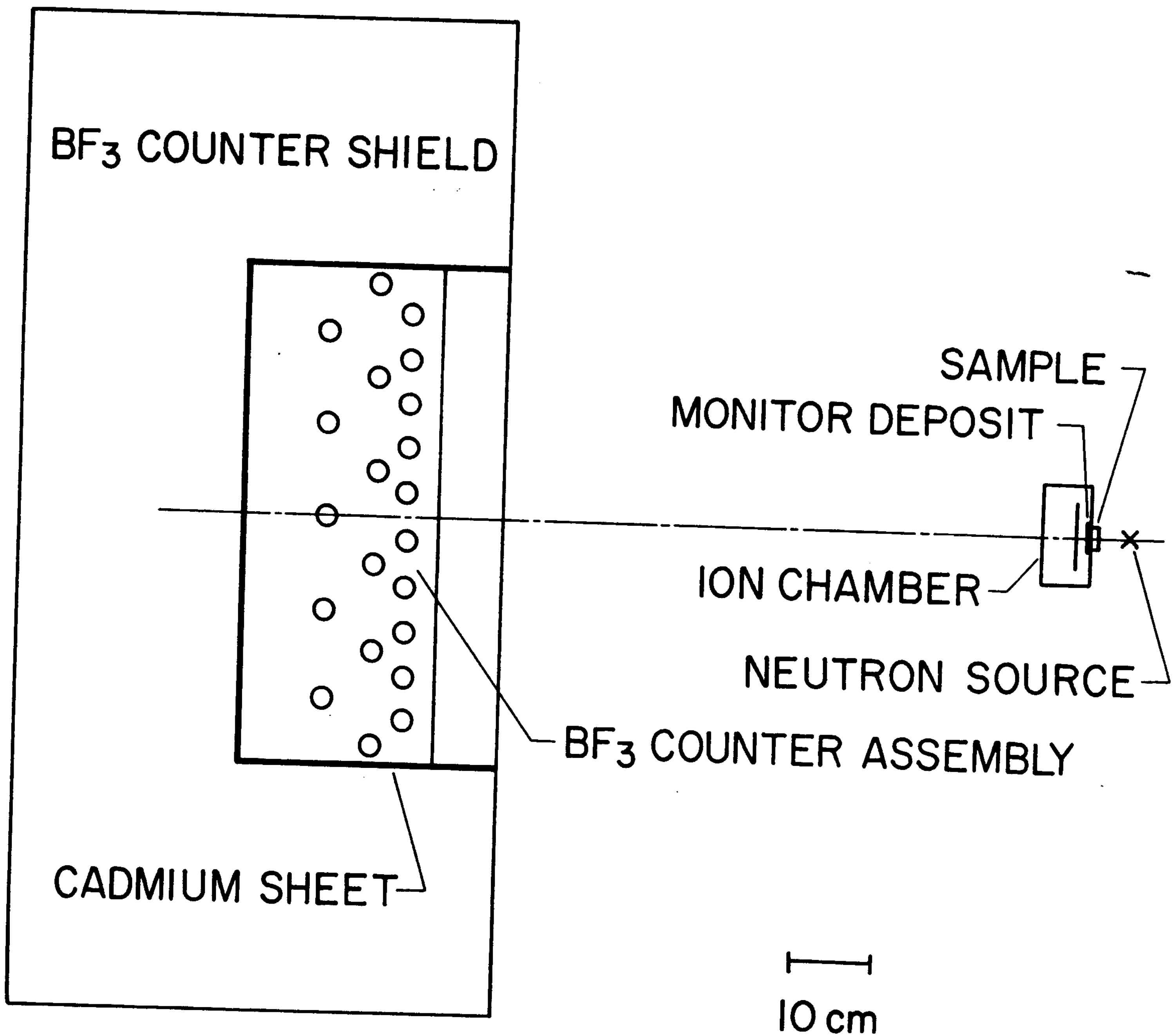

BF3 COUNTER SHIELD
SAMPLE
MONITOR DEPOSIT
ION CHAMBER
NEUTRON SOURCE
BF3 COUNTER ASSEMBLY
CADMIUM SHEET
10 cm

Fig. 2.